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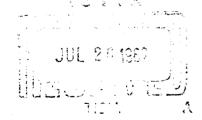
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ELECTRON DISTRIBUTION FUNCTION AND ELECTRICAL CONDUCTIVITY OF A SLIGHTLY IONIZED GAS

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ELECTRON DISTRIBUTION FUNCTION AND ELECTRICAL CONDUCTIVITY OF A SLIGHTLY IONIZED GAS

Ву

Douglas H. Sampson Jacob Enoch

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June 1962

MISSILE AND SPACE VEHICLE DEPARTMENT

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ABSTRACT

An expansion of a formally rigorous integral solution of the Boltzmann equation in powers of the electric field and is used to demonstrate clearly the correct expression for the conductivity of a spatially homogeneous slightly ionized gas. The conditions under which the isotropic part of the electron distribution function reduces to the Margenau distribution are determined. A discussion of the physical reasons for the possibility of negative conductivity is given.

I. INTRODUCTION

with electromagnetic fields as developed by many authors 1, 2, 3, 4, 5, 6 are usually based on a modified Lorentz gas 7 approximation. That is, electron-electron collisions as well as ion-ion collisions are ignored, and the recoil of the neutral molecules is taken into account to first order only. In addition the Boltzmann equation is expanded in Legendre polynomials in velocity space. The latter procedure leads to an infinite set of coupled differential equations for the expansion coefficients which is truncated, usually by setting all but the first two coefficients equal to zero. This procedure leads to the following expression for the current density:

$$j = \frac{4\pi e}{3} \int_0^{\infty} \langle \dot{x} \rangle v^3 = \frac{\partial f_0(v) dv}{\partial v} , \qquad (1)$$

in which (-e) is the charge of an electron and $f_0(v)$ is the isotropic part of the electron distribution function. For an electric field of the form

$$\epsilon(t) = \mathbf{E}\cos\boldsymbol{\omega} t \tag{2}$$

the quantity $\langle \dot{x} \rangle$ is given by the equation

$$\langle \dot{x} \rangle = \frac{-eE}{m(v^2 + \omega^2)} \left[v \cos \omega t + \omega \sin \omega t \right]$$
 (3)

where v is the electron collision frequency (normally a function of velocity).

In a later paper, 8 Margenau has shown that equation (1) is valid for more general conditions than initially had been demonstrated. It was then pointed out 9 that equation (1) does not in itself guarantee the positive definiteness of the electric conductivity, and that, in fact, substitution of a 8 -function in the magnitude of the electron velocity for $f_0(v)$ and the experimental collision frequency in air for v, yields a negative value for the conductivity. The following, alternate formula for the current density was proposed: 9

$$j = -4\pi e \int \langle \dot{x} \rangle v^2 f_0(v) dv$$
. (4)

In order to obtain this formula, which guarantees positive conductivities, the philosophy was adopted that the only effect of the electric field (which we take in the \times direction for definiteness) is to shift the "equilibrium distribution", $f_O(v)$ in the following fashion:

$$f(v_x, v_y, v_2, t) = f_0(v_x - g, v_y, v_2) (1 - \frac{\partial g}{\partial v_x}),$$
 (5)

where f(v, t) is the new distribution function, g is the velocity shift of an average electron, and the factor $(1 - \frac{\partial g}{\partial v_X})$ accounts for the fact that the volume element in the "shifted" region differs from the unshifted volume element. g was shown to be equal to $\langle \dot{x} \rangle$ as given by equation (3).

When the current density is calculated using a distribution function of the form given by equation (5) and is subsequently expanded in powers of $\langle \dot{x} \rangle$, the lowest order non-vanishing result is given by equation (4).

Taylor 10 has recently computed the average shift of the electron velocity by what is essentially a random walk technique. His result agrees with equation (3). Subsequently he uses equation (5) for the calculation of the current density to all orders in $\langle \dot{x} \rangle$ and, as a special case, obtains equation (4) to first order in $\langle \dot{x} \rangle$. Taylor's calculation of $\langle \dot{x} \rangle$, however, is correct only if the collision frequency is independent of velocity, because he neglects to take into account certain retardation effects, which we shall have occasion to discuss later on in this paper (equation (15)).

In Section II of this paper we expand a formally rigorous solution of the Boltzmann equation for a modified Lorentz gas in powers of the electric field and in powers of the ratio of the electron mass to the mass of the molecule. The function $f_0(v, t)$ is precisely defined and the

conditions under which it reduces to the steady state Margenau distribution are determined. In Section III the current density is computed and shown to be given by equation (1) to first order in the electric field. The physical reasons for the possibility of negative conductivities are discussed.

II. THE ELECTRON DISTRIBUTION FUNCTION

The Boltzmann equation for a modified Lorentz gas which is homogeneous in space and on which is imposed an electric field of the form given by equation (2) is

$$\frac{\partial f(\underline{v}, t)}{\partial t} + \gamma \cos \omega t \frac{\partial f(\underline{v}, t)}{\partial v_{x}} = -v(\underline{v}) f(\underline{v}, t) + S(\underline{v}, t)$$
 (6)

where $Y = \frac{-c}{m}$ E and S(v, t) is a source term. The collision frequency v(v) describes all types of collisions - elastic, inelastic, ionizing collisions, absorption, etc. S(v, t) is the rate at which the number of electrons in the velocity range v, v + dv is added due to elastic collisions as well as due to all other processes. In general S(v, t) depends on the form of f(v, t).

To obtain a formal solution of equation (6) we first make the following transformation:

$$v_x - \frac{y}{\omega} \sin \omega t = c$$
,

 $t - \frac{y}{\omega} t$. (7)

Let $F(c, v_y, v_z, t) = f(v_x, v_y, v_z, t)$, then the equation becomes

and

$$\frac{\partial F(c, v_y, v_z, t)}{\partial t} + v(c + \frac{y}{\omega} \sin \omega t, v_y, v_z) F(c, v_y, v_z, t) = S(c + \frac{y}{\omega} \sin \omega t, v_y, v_z).$$
(8)

The solution of this equation is:

$$F(c, v_{y}, v_{z}, t) = \int_{c}^{t} S(c + \frac{\gamma}{\omega} \sin t', v_{y}, v_{z}, t') \exp \left[- \int_{t'}^{t} v(c + \frac{\gamma}{\omega} \sin t'', v_{y}, v_{z}) dt'' \right] dt' + F(c, v_{y}, v_{z}, t_{o}) \exp \left[- \int_{t}^{t} v(c + \frac{\gamma}{\omega} \sin t', v_{y}, v_{z}) dt' \right],$$
(9)

or, after transformation to the original variables:

$$f(v_{x}, v_{y}, v_{z}, t) = \int_{t_{0}}^{t} S(v_{x} - \dot{x}(t, t'), v_{y}, v_{z}, t') \exp \left[- \int_{t'}^{t} \nu(v_{x} - \frac{\gamma}{\omega} \dot{x}(t, t''), v_{y}, v_{z}) dt'' \right] dt',$$

$$+ \exp \left[- \int_{t_{0}}^{t} \nu(v_{x} - \dot{x}(t, t'), v_{y}, v_{z}, t') dt' \right] f(v_{x} - \dot{x}(t, t_{0}), v_{y}, v_{z}, t_{0})$$
(10)

where the notation

$$\hat{\mathbf{x}}(\mathbf{t}, \, \mathbf{t}') = \frac{\mathbf{y}}{\omega} \left[\sin \omega \mathbf{t} - \sin \omega \mathbf{t}' \right] \tag{11}$$

has been introduced, and where t_0 is some initial time at which f is specified. Equation (10) is, of course, not a solution of the problem since S(v, t) is itself related to f(v, t). It is merely an integral form of equation (6) and is completely equivalent to it. Equation (10) does, however provide

a convenient starting point for a systematic computation of the distribution function as a series in powers of γ .

Consider the case where $t=-\infty$. Because of the positive definiteness of the collision frequency, ν , the second term on the right had side vanishes for all $t>-\infty$, leading to the equation

$$f(\mathbf{v}_{x}, \mathbf{v}_{y}, \mathbf{v}_{z}, t) = \int_{-\infty}^{t} S(\mathbf{v}_{x} - \dot{\mathbf{x}}(t, t'), \mathbf{v}_{y}, \mathbf{v}_{z}, t') \exp \left\{ -\int_{t'}^{t} \mathbf{v} \left[\mathbf{v}_{x} - \dot{\mathbf{x}}(t, t''), \mathbf{v}_{y}, \mathbf{v}_{z} \right] dt'' \right\} dt' .$$
(12)

We digress temporarily to discuss certain aspects of Taylor's 10 paper. Assume that the molecules are infinitely heavy and that the scattering cross section is isotropic. In this case

$$S(\underline{v}, t) = N \int_{\Omega} v'f(\underline{v}', t) \sigma(\underline{v}' \longrightarrow \underline{v}) d\Omega \left(\frac{d^3 v'}{d^3 v}\right)$$

$$= \nu(v) f_{\Omega}(v, t) = S_{\Omega}(v, t)$$
(13)

where $f_{O}(v, t)$ and $S_{O}(v, t)$ are defined by the equations

$$f_o(v, t) = \frac{1}{4\pi} \int_{\Omega} f(\underline{v}, t) d\Omega ; S_o(v, t) = \frac{1}{4\pi} \int_{\Omega} S(v, t) d\Omega .$$
 (14)

Here N is the number of molecules per unit volume and $\sigma(v \longrightarrow v)d\Omega$ is the differential scattering cross section. The final form of equation (13) results because in this particular case v' = v and σ is a function of the

velocity magnitude alone. We now multiply equation (12) by v_x , integrate over all velocities and divide by the electron density n to obtain the average electron velocity. After substitution for S(v, t) from equation (13), we obtain

$$\langle v_{x} \rangle = \frac{1}{n} \int_{-\infty}^{t} v_{x} \nu(v_{x} - \dot{x}(t, t'), v_{y}, v_{z}, t') f_{o}(v_{x} - \dot{x}(t, t'), v_{y}, v_{z}, t')$$

$$+ \exp \left\{ - \int_{t'}^{t} v \left[v_{x} - x(t, t''), v_{y}, v_{z} \right] dt'' \right\} dt' d^{3}v.$$
(15)

For the special case of velocity independent collision frequency and time independent f_0 , equation (15) reduces to

$$\langle v_{x} \rangle = \int_{-\infty}^{t} \dot{x}(t, t') \nu e^{-\nu (t - t')} dt'$$
 (16)

which upon slight change in notation reduces to equation (5) of Taylor's paper, and the current density is a linear function of the electric field regardless of the strength of the field. In general, as can be seen, a velocity dependence of ν implies a complicated time dependence of ν in integrals such as (16). If equation (15) is expanded in powers of γ (again, with isotropic, time independent f_0) $\langle v_\chi \rangle$ is not given by equation (16) but contains an additional term of the same order, which is proportional to $\frac{\partial \nu}{\partial v_\chi}$, and higher order terms in γ . This equation is therefore not adequate for the calculation of the conductivity even to lowest

order non-vanishing terms in γ , much less for the computation of higher order terms as Taylor has done.

We now return to the computation of the distribution function.

We expand equation (12) in powers of Y to obtain

$$f(\underline{v}, t) = \int_{-\infty}^{t} e^{-v(t-t')} dt' \left(S(\underline{v}, t') - \frac{\gamma}{\omega} \left[a_1(t, t') \frac{\partial S(\underline{v}, t')}{\partial v_X} - a_2(t, t') v^{-1} \frac{\partial v}{\partial v_X} S(\underline{v}, t') \right] + \frac{\gamma^2}{2\omega^2} \left\{ \left[\frac{a_2(t, t')}{v} \frac{\partial v}{\partial v_X} \right]^2 S(\underline{v}, t') - \frac{a_3^2(t, t')}{v} \frac{\partial^2 v}{\partial v_X^2} S(\underline{v}, t') \right\} - \frac{2a_1(t, t')a_2(t, t')}{v} \frac{\partial v}{\partial v_X} \frac{\partial S(v, t')}{\partial v_X} + a_1^2(t, t') \frac{\partial^2 S(v, t')}{\partial v_X^2} \right\} + \dots$$

where V = V(V) and

$$a_{1}(t, t') = \frac{\omega}{\gamma} \dot{x}(t, t'); a_{2} = \frac{\omega \nu}{\gamma} \int_{t'}^{t} \dot{x}(t, t'')dt''$$

$$a_{3}^{2}(t, t') = \frac{\omega^{2} \nu}{\gamma^{2}} \int_{t'}^{t} \dot{x}^{2}(t, t'')dt''.$$
(18)

We now integrate both sides of equation (17) over solid angle in velocity space, and obtain, under the assumption that S(v, t) is isotropic, the following equation:

$$f_{o}(v, t) = \int_{-\infty}^{t} e^{-v(t-t')} dt' \left(S_{o}(v, t') + \frac{\gamma^{2}}{6\omega^{2}} \left\{ S_{o}(v, t') \left[a_{2}^{2}(t, t') \left(\frac{1}{v} \frac{dv}{dv} \right)^{2} - \frac{a_{3}^{2}(t, t')}{v} \left(\frac{2}{v} \frac{dv}{dv} + \frac{d^{2}v}{dv^{2}} \right) \right] - \frac{2}{v} a_{1}(t, t') a_{2}(t, t') \frac{dv}{dv} \frac{\partial S_{o}(v, t')}{\partial v} + a_{1}^{2}(t, t') \left(\frac{2}{v} \frac{\partial}{\partial v} + \frac{\partial^{2}v}{\partial v^{2}} \right) S_{o}(v, t') \right\} + O(\gamma^{4}) .$$
(19)

Then we differentiate equation (19) and get:

$$\frac{\vartheta f_{o}(v, t)}{\vartheta t} = -v f_{o}(v, t) + S_{o}(v, t) + \frac{\gamma^{2}}{6\omega^{2}} \int_{-\infty}^{t} e^{-v (t - t^{1})} dt^{1}$$

$$\cdot \left\{ S_{o}(v, t^{1}) \left[\left(\frac{1}{v} \frac{dv}{dv} \right)^{2} \frac{\vartheta a_{2}^{2}(t, t^{1})}{\vartheta t} - \frac{1}{v} \left(\frac{2}{v} \frac{dv}{dv} + \frac{d^{2}v}{dv^{2}} \right) \frac{\vartheta a_{3}^{2}(t, t^{1})}{\vartheta t} \right] \right.$$

$$\left. - \frac{2}{v} \frac{dv}{dv} \frac{\vartheta S_{o}(v, t^{1})}{\vartheta v} \frac{\vartheta}{\vartheta t} \left[a_{1}(t, t^{1}) a_{2}(t, t^{1}) \right] + \left[\left(\frac{2}{v} \frac{\vartheta}{\vartheta v} + \frac{\vartheta^{2}}{\vartheta v^{2}} \right) S_{o}(v, t^{1}) \right] \right.$$

$$\left. \cdot \frac{\vartheta a_{1}^{2}(t, t^{1})}{\vartheta t} \right\} + O(\gamma^{4}). \tag{20}$$

With the exception of a slight digression to a discussion of more general conditions following equation (49) in Section III, we shall restrict ourselves to the case where elastic scattering of electrons by neutral molecules is the only contribution to the source term for the remainder of this paper. Also, with the exception of the appendix, we consider only the case where the differential scattering cross section is isotropic in the

center of mass system. Note that for infinitely heavy molecules, these assumptions imply that S(v, t) has the isotropic value given by equation (13). Substitution from this relationship into equation (20) shows that $\frac{\partial f_0}{\partial t}$ is small of order γ^2 .

We now examine the corrections to equation (13) which arise when recoil is taken into account to first order.

Let $\sigma(v', \theta) d\Omega$ be the differential cross section in the laboratory system for scattering electrons through an angle θ into the velocity range v, v + dv. We assume that the temperature of the neutral molecules is low enough so that their velocity before the collision can be ignored; thus v' is the velocity of the electron before the collision and θ is the scattering angle in the <u>laboratory frame of reference</u>. As a consequence of the invariance of the differential scattering cross section to a transformation to the center of mass system we have

$$\sigma(v', \theta) d\Omega = \sigma_c(g, \theta_c) d\Omega_c$$
 (21)

in which the subscript "c" refers to center of mass system and g is the magnitude of the relative velocity. Using the relations resulting from conservation of energy and momentum and the fact that the velocity of the molecule before collision is taken as zero, we get the equation

$$\cos\theta_{c} = \cos\theta + \frac{m}{M} (\cos^{2}\theta - 1) + O(\frac{m}{M})^{2}, \qquad (22)$$

where m and M are the electron and molecule masses, respectively.

The azimuthal angle is invariant to the transformation. Thus

$$\sigma(v', \theta) = \sigma_c(g, \theta_c) \frac{d(\cos \theta_c)}{d(\cos \theta)} = (1 + 2 \frac{m}{M} \cos \theta) \sigma_c(v', \theta_c), \quad (23)$$

where we used the relation v' = g in the last expression. In general, assuming $\sigma_c(g, \theta_c)$ is known, one would also use equation (22) to substitute for θ_c . In the case considered here the cross section is isotropic in the center of mass system. Thus

$$\sigma(\mathbf{v}^{i}, \boldsymbol{\theta}) = (1 + 2 \frac{\mathbf{m}}{\mathbf{M}} \cos \boldsymbol{\theta}) \ \sigma_{\mathbf{c}}(\mathbf{v}^{i}) + O(\frac{\mathbf{m}}{\mathbf{M}})^{2} \ . \tag{24}$$

Following Morse, Allis, and Lamar², we get the following form for the source function:

$$S(\underline{v}, t) = \frac{N}{v^3} \int_{\Omega} f(\underline{v}', t) (v')^4 \sigma(v', \theta) d\Omega$$
 (25)

where N is the number of molecules per unit volume. In the integration of equation (25) over solid angle \underline{v} rather than \underline{v}' is held constant. Thus one must express v' in terms of v and θ . The required relation is

easily obtained by using the equations expressing conservation of momentum and energy. The result is

$$v' = v \left[1 + \frac{m}{M} \left(1 - \cos \theta \right) \right] + O\left(\frac{m}{M} \right)^2.$$
 (26)

We now split the distribution function into isotropic and non-isotropic parts,

$$f(v, t) = f_0(v, t) + f_1(v, t)$$
 (27)

and initially make no assumptions as to the relative size of the two parts.

Using equations (24), (26) and (27) in equation (25) and expanding the result in powers of m/M we obtain

$$S(\underline{v}, t) = v(v)f_{0}(v, t) + \frac{m}{Mv^{2}} \frac{\vartheta}{\vartheta v} \left[v^{3}v(v)f_{0}(v, t) \right]$$

$$+ \frac{2m}{M} v(v)f_{1}^{1}(\underline{v}, t) - \frac{m}{Mv^{2}} \frac{\vartheta}{\vartheta v} \left[v^{3}v(v)f_{1}^{1}(\underline{v}, t) \right] + O(\frac{m}{M})^{2}$$

$$(28)$$

where

$$T_1^{1}(v, t) = \int_{\Omega} f(v', t) \cos \theta d\Omega, \quad v(v) = N \int_{\Omega} v \sigma(v, \theta) d\Omega = 4\pi N v \sigma_c(v) + O(\frac{m}{M})^2.$$
(29)

Consider the integral expression for f_1^{-1} . It can be written as

$$f_1^1 = \int_{\Omega} f(\underline{y}^1, t) \frac{\underline{y}^1 \cdot \underline{y}}{\underline{y}^1 \cdot \underline{y}} d\Omega$$

and, since $v' = v + O(\frac{m}{M})$ one can write

$$f_1^{-1} = \int f(\underline{v}^i, t)\underline{v}^i \cdot \underline{v} \frac{\delta(v^i + v)}{v^4} d^3v^i + O(\underline{m}) .$$

Clearly $f_1^{-1}(\underline{v}, t)$ is an odd function of the components of \underline{v} to order $\frac{m}{M}$, and is hence, in this approximation, purely anisotropic, that is

$$\int f_1^{-1} d\Omega = O(\frac{m}{M}) . \tag{30}$$

When S(v, t), as given by equation (28) is substituted on the right hand side of equation (17) it is seen that f(v, t) is isotropic if terms of order γ and higher order terms are neglected. That is $f_1(v, t)$ is of order γ . By making use of this fact we can rewrite equation (28) as follows.

$$S(\underline{v}, t) = v(v)f_{0}(v, t) + \frac{m}{Mv^{2}} \frac{3}{3v} \left[v^{3}v(v)f_{0}(v, t)\right] + O(\frac{m}{M})^{2} + \text{purely anisotropic terms of order } \gamma \frac{m}{M}.$$
(31)

We can thus see that when the center of mass differential scattering cross section is isotropic, the source term is isotropic through order $\frac{m}{M}$.

In the derivation of equation (19) and (20) we have assumed that the source is isotropic. By substituting S(v, t) as given by equation (31) into equation (17) and repeating the ensuing calculation, (without the explicit assumption that S(v, t) is purely isotropic), we see that equations (19) and (20) are still satisfied if terms of order $\gamma \frac{2m}{M}$ and higher order terms are neglected. In particular equation (20) takes on the following form:

$$\frac{\partial f_{O}(v, t)}{\partial t} = \frac{m}{Mv^{2}} \frac{\partial}{\partial v} \left[v^{3} v f_{O}(v, t) \right] + \frac{\gamma^{2}}{6 \omega^{2}} \int_{-\infty}^{t} e^{-v(t-t')} v dt' \\
\cdot \left(f_{O}(v, t') \left\{ \left(\frac{1}{v} \frac{dv}{dv} \right)^{2} \frac{\partial}{\partial t} \left[a_{2}^{2}(t, t') - 2a_{1}(t, t') a_{2}(t, t') \right] \right. \\
+ \frac{1}{v} \left(\frac{2}{v} \frac{dv}{dv} + \frac{d^{2}v}{dv^{2}} \right) \frac{\partial}{\partial t} \left[a_{1}^{2}(t, t') - a_{3}^{2}(t, t') \right] \right\} \\
+ \frac{2}{v} \frac{dv}{dv} \frac{\partial f_{O}(v, t')}{\partial v} \frac{\partial}{\partial t} \left[a_{1}^{2}(t, t') - a_{1}(t, t') a_{2}(t, t') \right] \\
+ \left[\left(\frac{2}{v} \frac{\partial}{\partial v} + \frac{\partial^{2}}{\partial v^{2}} \right) f_{O}(v, t') \right] \left(\frac{\partial a_{1}^{2}(t, t')}{\partial t} \right) + O(\gamma^{2} \frac{m}{M}) + O(\gamma^{4})$$

Equation (32) implies that $f_0(v, t)$ is a rather weak function of time ($\frac{\theta f_0}{\theta t}$ is of order $\frac{m}{M}$ or y^2 , whichever is larger). This fact, together with the fact that the presence of the exponential $e^{-\nu(\frac{1}{t}-t')}$ suppresses contributions with t' << t in the integral of the right hand side

of equation (32), suggests that we can substitute $f_O(v, t)$ for $f_O(v, t')$ under this integral. When the indicated integration is performed it turns out that the part in the braces contributes nothing. The result is

$$\frac{\partial f_{O}(v, t)}{\partial t} = \frac{m}{Mv^{2}} \frac{\partial}{\partial v} \left[v^{3} v f_{O}(v, t) \right]$$

$$+ \frac{v^{2}}{6 \omega^{2}} \left[A_{1}(t) \left(\frac{2}{v} \frac{\partial}{\partial v} + \frac{\partial^{2}}{\partial v^{2}} \right) f_{O}(v, t) + \frac{2A_{2}(t)}{v} \frac{\partial v}{\partial v} \frac{\partial f_{O}(v, t)}{\partial v} \right]$$

$$+ O(v^{2} \frac{m}{M}) + O(v^{4}).$$
(33)

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$$A_{1}(t) = \int_{-\infty}^{t} e^{-v(t-t')} dt' \frac{\partial}{\partial t} a_{1}^{2}(t, t') = \frac{2\omega^{2}}{v^{2} + \omega^{2}} (\cos^{2}\omega t + \frac{\omega}{v} \sin \omega t \cos \omega t);$$

and

$$A_{2}(t) = \int_{-\infty}^{t} e^{-v(t-t')} dt' \frac{\partial}{\partial t} \left[a_{1}^{2}(t, t') - a_{1}(t, t') a_{2}(t, t') \right]$$

$$= \frac{\omega^{2}}{(v^{2} + \omega^{2})^{2}} \left[(\omega^{2} - v^{2}) \cos^{2} \omega t - 2 \omega v \sin \omega t \cos \omega t \right]$$
(34)

where $a_1(t, t')$ and $a_2(t, t')$ are given by equations (11) and (18). Substituting the values given by equation (34) into equation (33) and rearranging the

term proportional to γ^2 we get

$$\frac{\partial f_{O}(v, t)}{\partial t} = \frac{m}{Mv^{2}} \frac{\partial}{\partial v} \left[v^{3} v f_{O}(v, t) \right]$$

$$+ \frac{\gamma^{2}}{3v^{2}} \frac{\partial}{\partial v} \left[\left(\cos^{2} \omega t + \frac{\omega}{v} \sin \omega t \cos \omega t \right) \frac{v^{2}}{\omega^{2} + v^{2}} \frac{\partial f_{O}(v, t)}{\partial v} \right] + O(\gamma^{2} \frac{m}{M}) + O(\gamma^{4})$$

By making use of the weak time dependence of $f_0(v, t)$, we can integrate over a period of the electric field holding $f_0(v, t)$ constant. Furthermore, if we add to the term proportional to m/M the well-known correction due to the thermal motion of the molecules, $f_0(v, t)$, we get the steady state result

$$-\frac{y^2}{6} \frac{d}{dv} \left[\frac{v^2 v}{\omega^2 + v^2} \frac{df_0(v)}{dv} \right] = \frac{m}{M} \frac{d}{dv} \left\{ v^3 v \left[f_0(v) + \frac{kT}{mv} \frac{df_0(v)}{dv} \right] \right\}$$
(36)

which upon integration leads to the Margenau distribution function 6

$$f_{O}(v, t) = f_{O}(v) = C \exp \left[-\int_{0}^{v} \frac{m v dv}{kT + y^{2} M/6(\omega^{2} + v^{2})} \right],$$
 (37)

in which C is a normalization constant.

Once again we make use of the result that the time dependence of $f_0(v, t)$ is insignificant and integrate equation (33) over time, holding $f_0(v, t)$ fixed on the right hand side. The result can be used to express $f_0(v, t)$ in terms of $f_0(v, t)$.

$$f_{o}(v, t') = f_{o}(v, t) + (t' - t) \frac{m}{Mv^{2}} \frac{\partial}{\partial v} \left[v^{3} v f_{o}(v, t) \right]$$

$$- \frac{v v^{2}}{6 \omega^{2}} \left\{ \left[\int_{t'}^{t} A_{1}(t'') dt'' \right] \left(\frac{2}{v} \frac{\partial}{\partial v} + \frac{\partial^{2}}{\partial v^{2}} \right) f_{o}(v, t) + \left[\int_{t'}^{t} A_{2}(t'') dt'' \right] \left(\frac{2}{v} \frac{\partial v}{\partial v} - \frac{\partial f_{o}(v, t)}{\partial v} \right) \right\} + \dots$$

$$(38)$$

Now, by using this equation together with equation (31) we obtain

$$S(\underline{v}, t') = \nu f_0(v, t) + \left[1 - \nu(t - t')\right] \frac{m}{Mv^2} \frac{\partial}{\partial v} \left[v^3 \nu f_0(v, t)\right] - \frac{v^2 y^2}{6\omega^2} \left\{\right\},$$
(39)

where the curly brackets are meant to contain the same quantity as they do in equation (38). We substitute the r.h.s. of equation (39) for S(v, t') in the zeroth order term of equation (17), while the value $vf_0(v, t)$ is used for S(v, t') in the higher order terms. The term proportional to m/M and the terms proportional to $\frac{3\nu}{3v_x}$. $\frac{3\nu}{3v_x}$, or $(\frac{3^2\nu}{3v_x})^2$, times $f_0(v, t)$ drop out in the integration over t and we get

$$f(v, t) = f_0(v, t) - \frac{\gamma}{\omega} b(t) \frac{v_x}{v} \frac{\partial f_0(v, t)}{\partial v}$$

$$+ \frac{\gamma^2}{2\omega^2} \left(\frac{v_x^2}{v^2} - \frac{1}{3} \right) \left\{ B_1(t) \left(-\frac{1}{v} \frac{\partial}{\partial v} + \frac{\partial^2}{\partial v^2} \right) f_0(v, t) + \frac{2B_2(t)}{v} \frac{\partial v}{\partial v} \frac{\partial f_0(v, t)}{\partial v} \right\} + O(\gamma \frac{m}{M}) + O(\gamma^3)$$
(40)

(41)

where

$$b(t) = \int_{-\infty}^{t} e^{-\nu(t-t')} \nu dt' a_1(t, t') = \frac{\omega}{\nu^2 + \omega^2} \left[\nu \cos \omega t + \omega \sin \omega t \right]$$

$$B_1(t) = \int_{-\infty}^{t} e^{-\nu(t-t')} \nu dt' \left[a_1^2(t, t') \right] = \frac{\omega^2}{(\nu^2 + \omega^2)} \left[1 + \frac{3\omega \nu \sin 2\omega t + (\nu^2 - 2\omega^2)\cos 2\omega t}{(\nu^2 + 4\omega^2)} \right],$$

and

$$B_{2}(t) = \int_{-\infty}^{t} e^{-v(t-t')} v dt' \left[a_{1}^{2}(t, t') - a_{1}(t, t') a_{2}(t, t') \right]$$

$$= \frac{\omega^{2}}{2(v^{2} + \omega^{2})} \left[\frac{\omega^{2} - v^{2}}{\omega^{2} + v^{2}} + \frac{(2\omega^{3}v - 4v^{3}\omega)\sin2\omega t + (5\omega^{2}v^{2} - v^{4})\cos2\omega t}{(\omega^{2} + v^{2})(v^{2} + 4\omega^{2})} \right]$$

In obtaining equation (40) we made use of the fact that $B_1(t)$ and $B_2(t)$ can be expressed as follows:

$$B_{1}(t) = \int_{-\infty}^{t} e^{-\sqrt{(t-t')}} y^{2} dt' \left[\int_{t'}^{t} A_{1}(t'') dt'' \right], \quad B_{2}(t) = \int_{-\infty}^{t} e^{-\sqrt{(t-t')}} y^{2} dt' \left[\int_{t'}^{t} A_{2}(t'') dt'' \right].$$
(42)

We see that our solution for the electron distribution written as an expansion in powers of the electric field turns out also to be an expansion in Legendre polynomials in the cosine of the angle between \underline{v} and the electric field to the order for which we have determined the coefficients of the expansion.

In the conventional treatment the distribution function is expanded in spherical harmonics at the outset and the assumption is made that the contributions

proportional to Legendre polynomials of second order and higher are negligible. The justification for this assumption being that when it is made the series appears to converge rapidly on the basis of the two or three terms which are computed.

Although our method is lengthy, it seems to us to be more satisfactory because we have shown that the contributions not explicitly given in equation (40) are indeed of higher order in terms of certain expansion parameters (γ and m/M) than those retained. Furthermore, we have explicitly demonstrated that the oscillating portion is indeed small.

III. ELECTRICAL CONDUCTIVITY

If we substitute the result given by equation (40) into the expression for the current density,

$$j = -e \int v f(v, t) d^3v , \qquad (43)$$

we obtain a result in agreement with equations (1) and (3)

$$j = -\frac{4\pi}{3} \frac{e^2 E}{m} \int_0^{\infty} \left(\frac{v \cos \omega t + \omega \sin \omega t}{\omega^2 + v^2} \right) v^3 \frac{\partial f_0(v, t) dv}{\partial v} + O(\gamma \frac{m}{M}) + O(\gamma^3).$$
(44)

Thus for elastic scattering which is isotropic in the center of mass system we have proven that equation (1) rather than equation (4) is the correct expression for the current density; however, equation (1) is valid for much more general conditions. In fact it is valid to lowest order in powers of γ when the source S(v, t) is due to any processes provided that the lowest order anisotropy in S(v, t) is of order γ^2 or higher and that the time dependence of S(v, t) is of order γ or higher. From equation (19) we see that the latter condition implies that the purely isotropic part of S(v, t) is equal to $\nu f_0(v, t)$ plus terms of order γ or higher. Thus the lowest order contribution to equation (43) for the conductivity results from substitution of $\nu f_0(v, t)$ for S(v, t') in the term proportional to γ in equation (17) for f(v, t). As noted in obtaining equation (40), the terms proportional to $\nu f_0(v, t)$

the integration over t' and with the aid of the first of equations (41) one obtains equation (44) with corrections of order γ^2 or higher for the current density.

In order to demonstrate clearly the velocity dependence of f_0 and ν required for negative conductivity, we integrate equation (44) by parts, obtaining

$$j = -\frac{4\pi}{3} \frac{e^2 E}{m} \left[\frac{(v\cos\omega t + \omega\sin\omega t)}{\omega^2 + v^2} v^3 f_0(v, t) \right]_0^\infty + \frac{4\pi}{3} \frac{e^2 E}{m} \int_0^\infty f_0(v, t) dv \frac{d}{dv} \left[\frac{v^3 (v\cos\omega t + \omega\sin\omega t)}{v^2 + \omega^2} \right].$$

For any finite ω the first term must vanish in order that the energy density of the electrons be finite. We take the scalar product of j with the electric field, average over a period of the field, and obtain the average power given by the field to the electrons

$$\vec{p} = \frac{4\pi}{3} - y^2 m \int_0^{\infty} f_0(v, t) dv \left\{ \frac{3v^2 v}{2(v^2 + \omega^2)} + \frac{v^3(\omega^2 - v^2)}{2(v^2 + \omega^2)^2} \frac{dv}{dv} \right\}$$
(45)

In the vicinity of a particular speed v_1 we approximate the collision frequency by $\mathcal{V}(v_1) = av_1^n$. Then it is apparent that for $\omega >> \mathcal{V}(v_1)$ electrons of speed v_1 lose energy to the field (in phase part of the conductivity negative) if n < -3, while for $\omega << \mathcal{V}(v_1)$ they lose energy to the field if n > 3. Thus if the distribution function is sharply peaked about the speed v_1 for

which $v(v_1)$ satisfies either of these conditions, the total power gained by the electrons from the field is negative.

It is possible to give a rough, qualitative, explanation of this negative conductivity phenomenon. In the case where $\omega >> y$ and y decreases with velocity, the electron oscillates about its initial velocity many times between collisions and the probability that it will undergo a collision during the portion of the cycle during which its speed is below its initial speed is higher than the probability that it will collide during the portion of cycle when its speed is above its initial speed. It will thus on the average lose energy to the field. In the case where $\omega << y$ and y increases with velocity it is easiest to think of the situation as approximating the D. C. Case. After a collision an electron may move either with or against the field (with equal probability for isotropic scattering). Since the collision frequency increases with speed, the electrons which are being decelerated will collide later, on the average, than those that are being sccelerated. Thus electrons spend more time giving energy up to the field than gaining energy from it.

Since we integrated from -& to t in obtaining equations (40) and (44) one might expect our equation (44) to be valid only when $f_Q(v, t)$ is equal to the steady state distribution (the Margenau Distribution) given by equation (37). When this distribution function is used in equation (44) positive values result for the conductivity, regardless of the velocity dependence of V. Actually, if initially the electrons have some strange, non-isotropic,

distribution, equations (40) and (44) become applicable after a time equal to only a few multiples of the collision time y-1. Physically the time required is the time necessary for nearly all electrons to have suffered one or more collision, mathematically it is the time t required for transient terms proportional to exp (- Vt) to become negligible. This time is really very small in comparison with the time required to reach a steady state. A steady state is reached when the average energy gained by an electron from the field between successive collisions is equal to the average energy lost by an electron to a molecule per collision. Both these energies are small compared with the average kinetic energy per electron. The condition that the energy gained between collisions be small compared with the average kinetic energy per electron must be met in order for our approach to be valid (a condition met by most physical cases). The ratio of energy loss per collision to electron kinetic energy is $\sim m/M \sim 10^{-5}$. Thus we expect that a time equal to thousands of collision times may be required before a steady state is reached. As an example suppose that initially a neutral gas in a fairly weak electric field is bombarded with a beam of low energy electrons. After a few collision times the electron distribution is changed from an approximate delta function of velocity to an approximate delta function of energy and equations (40), (44) and (45) apply. If the velocity dependence of V(v) happens to meet the conditions discussed below equation (45), energy will be lost by the electrons to the field, perhaps for a detectable length of time if the density of the gas is low (y small), As the steady state is approached the distribution becomes peaked about the origin in velocity space and the conductivity becomes positive as seen from equation (44).

We turn now to a consideration of the contribution of higher order terms in powers of γ . These terms might be expected to give significant contributions to equations (37), (40) and (44) in the case of strong fields. In this connection we will also discuss more fully the criteria for the validity of the expansion in powers of γ used in obtaining these equations.

In principle our technique can be used to obtain the part of f(v, t) which is proportional to γ^3 ; however, it would be rather tedious. Instead we show by a more careful consideration of equation (37) that such a calculation would be of little value because for steady state conditions the contribution of the next higher order terms in powers of γ^2 to either $f_0(v, t)$ or j are of order m/M or less compared to those retained in equations (37) and (44).

We see this in the following way: if the second term in the denominator of the exponential in equation (37) is large compared with the first we have

$$\frac{M\gamma^2}{6(\omega^2 + v^2)} \simeq kT_e \simeq \frac{m\overline{v}^2}{2}$$
 (46)

or

$$\frac{y^2}{(\omega + y)\overline{v}} \simeq \frac{m}{M}$$
(47)

where T_e is the effective electron temperature obtained by setting kT_e equal to the denominator of the exponent of equation (37) and ∇ is the approximate average magnitude of v. On the other hand, if $kT > \gamma^2 M(\omega^2 + \gamma^2)^{-1}$ we see that $\gamma^2(\nabla)^{-2}(\omega^2 + \gamma^2)^{-1}$ is even smaller. The criterion for the validity of equation (40) is, of course, that the ratio of successive terms be less than unity. From inspection of equations (40) and (41) this criterion is roughly

$$\frac{\gamma}{\omega \nabla} < 1; \; \omega > \nu$$

and (48)

$$\frac{y}{y\nabla}$$
 < 1, $\omega < y$

comparing this with equation (47) we see that in the steady state condition a slightly ionized gas adjusts itself automatically in such a way that the expansion given by equation (40) is valid and the higher order corrections to equations (37) and (44) are of order m/M or less relative to those retained. Of course, for high enough field intensities kT_e becomes so large that various kinds of inelastic and ionizing collisions become frequent.

Then our assumption that all collisions other than elastic collisions between electrons and molecules are insignificant in determining f(v, t) breaks down. Thus the only circumstances for which terms of order y^3 could give a significant contribution to equation (44) would be for short times after an electric field was applied to a slightly ionized gas in which the electrons initially had energies of the same order or less than that gained on the average by an electron from the field between successive collisions. For constant collision frequency and isotropic distribution of electrons the latter energy is $(\omega^2 + \psi^2)^{-1} y^2 m/2$. Thus, it is interesting to note that the criteria given by the inequalities (48) for the validity of our expansion in powers of y is roughly equivalent to demanding that the average energy gained by an electron from the field between collisions (assuming y is not a very strong function of v) be small compared with the average kinetic energy per electron.

It is desirable to have the results for the case of non-isotropic scattering in the center of mass system as well. The conventional result for this case is that equation (37) and (44) apply with V replaced with V_m , the collision frequency for momentum transfer. This problem is considered in the appendix. There it is shown what conditions must be met by the part of f(v, t) proportional to Legendre Polynomials $P_{f}(v_x/v)$ with $f \ge 2$ in order to satisfy the requirements for the validity of the conventional result. It is plausible that these conditions are met, at least for the steady state; however, in contrast to the case of isotropic scattering, we are unable to show rigorously that they are met.

APPENDIX

It is our purpose here to generalize to the case of non-isotropic scattering. In this case we define a pseudo source function $S_m(v,t)$ by the equation

where v_{m} is the collision frequency for momentum transfer

$$\nu_{\rm m}({\bf v}) = N{\bf v} \int_{\Omega} (1 - \cos \theta) \, \sigma({\bf v}, \, \theta) d\Omega$$
 (50)

When equation (49) is used to express the right hand side of the Boltzmann equation, equation (6), we obtain the generalized versions of equations (12), (17) and (18). By the generalized version of an equation we will mean that the only change from the original is that V(v) and S(v, t) are everywhere replaced with $V_m(v)$ and $V_m(v, t)$. The symmetry properties of $V_m(v, t)$ can be investigated by solving equation (49) for V_m and by using equations (25) and (27) together with the definition of V_m , second of equations (29). We get

$$S_{m}(\underline{v}, t) = \frac{N}{v^{3}} \int_{\Omega} \left[f_{o}(v', t) + f_{1}(v', t) \right] (v')^{4} \sigma(v', \theta) d\Omega$$

$$- Nv \left[f_{o}(v, t) + f_{1}(\underline{v}, t) \right] \int_{\Omega} \sigma(v, \theta) \cos\theta d\Omega . \qquad (51)$$

The only agency producing asymmetries in the distribution function is the electric field. Thus the purely anisotropic part of the distribution function can be expanded in Legendre polynomials as follows:

$$f_1(v, t)^* = \int_{-1}^{\infty} C_{\ell}(v, t) P_{\ell}(\cos \psi); \cos \psi = v_{\chi}/v. \qquad (52)$$

Now we substitute this expression into equation (51) and expand functions of v'in powers of m/M about v with the use of equation (26). We obtain

$$S_{\mathbf{m}}(\underline{\mathbf{v}}, t) = \mathbf{v}_{\mathbf{m}} f_{\mathbf{o}}(\mathbf{v}, t) + N\mathbf{v} \int_{\mathbf{k}=1}^{\infty} C_{\mathbf{k}}(\mathbf{v}, t) \int_{\mathbf{\Omega}} \left[P_{\mathbf{k}}(\cos \psi^{\dagger}) - P_{\mathbf{k}}(\cos \psi) \cos \theta \right] \sigma(\mathbf{v}, \theta) d\Omega$$

$$+ \frac{m}{M\mathbf{v}^{2}} \frac{\partial}{\partial \mathbf{v}} \left\{ \mathbf{v}^{3} \mathbf{v}_{\mathbf{m}} f_{\mathbf{o}}(\mathbf{v}, t) + N\mathbf{v}^{4} \int_{\mathbf{k}=1}^{\infty} C_{\mathbf{k}}(\mathbf{v}) \int_{\mathbf{\Omega}} P_{\mathbf{k}}(\cos \psi^{\dagger}) (1 - \cos \theta) \sigma(\mathbf{v}, \theta) d\Omega \right\}.$$
(53)

We now make use of the well known relation

$$P_{\ell}(\cos\psi') = P_{\ell}(\cos\psi) P_{\ell}(\cos\theta) + 2\sum_{m=1}^{\ell} \frac{(\ell-m)!}{(\ell+m)!} P_{\ell}^{m}(\cos\psi) P_{\ell}^{m}(\cos\theta) \cos m(\phi - \phi_{2})$$
(54)

where $(\phi - \phi_2)$ is the angle between the projections of χ' and E in the plane perpendicular to χ . Since there is no azimuthal asymmetry in our problem, the part of equation (54) proportional to $cosm(\phi - \phi_2)$ gives no contribution when equation (54) is substituted into equation (53). Thus we obtain

$$S_{\mathbf{m}}(\mathbf{v}, t) = \mathbf{v}_{\mathbf{m}} f_{\mathbf{o}}(\mathbf{v}, t) + \sum_{k \geq 2}^{\infty} C_{k}(\mathbf{v}, t) P_{k}(\cos \psi) \left[\mathbf{v}_{\mathbf{m}}(\mathbf{v}) - \mathbf{v}_{k}(\mathbf{v}) \right] + O(\frac{\mathbf{m}}{M})$$
(55)

in which

$$\nu_{\ell}(v) = Nv \int_{\Omega} \left[1 - P_{\ell}(\cos\theta)\right] \sigma(v, \theta) d\Omega . \qquad (56)$$

In contrast to this result we recall that the source function for the case of isotropic scattering in the center of mass system had no zero order contribution in powers of m/M arising from $f_1(v, t)$, the anisotropic part of the distribution function. By our particular choice of definition for the pseudo source function $S_m(v, t)$ we have eliminated the zero order contribution proportional to $P_1(\cos \psi) = \cos \psi$. However, one sees that if $S_m(v, t)$ had been defined by equations (49) and (50) with any other Legendre polynomial $P_1(\cos \theta)$ replacing $\cos \theta$ in equation (50), then the zero order anisotropy proportional to $P_1(\cos \psi)$ would have been eliminated from $S_m(v, t)$. Nevertheless the choice made for the definition of $S_m(v, t)$ is most convenient.

If we make the assumption (usually made) that the part of f(v, t) proportional to Legendre polynomials $P_{\mathcal{C}}(\cos \psi)$ with $1 \ge 2$ is negligible, $S_{m}(v, t)$ is sufficiently isotropic for us to proceed as in Section II. We then get the conventional result that the steady state value of $f_{0}(v, t)$ is given by the generalized version of equation (37) and the current density by the generalized versions of equations (1) or (44). Actually all that is

required for these results to apply is that the C_{ℓ} (v, t) with $\ell \geq 2$ be of first order or higher in powers of γ . When the generalized version of equation (17) is integrated over angle to get the generalized versions of equations (19) and (20), the C_{ℓ} (v, t) with $\ell \geq 2$ appear only in terms proportional to γ^2 or m/M as a consequence of the orthogonality properties of Legendre polynomials. Thus, if all C_{ℓ} (v, t) with $\ell \geq 2$ are of order γ or higher, they contribute terms to equations (19) and (20) which are of order γ^3 and γ m/M or higher and can be neglected. We will show now that the requirement that the C_{ℓ} (v, t) be of order γ or higher for γ^3 reduces to the requirement that γ^2 times the approximate average change in C_{ℓ} (v, t) between collisions be of order γ or higher. An equation for C_{ℓ} (v, t) is obtained by substituting the value for the pseudo source function given by equation (55) into the generalized version of equation (17). Multiplying the equation through by γ^2 (cos γ^2), and integrating over solid angle, one gets

$$C_2(v, t) = (v_m - v_2) \int_{-\infty}^{t} e^{-v_m(t-t')} C_2(v, t') dt' + O(v^2)$$
 (57)

which can be rearranged as

$$C_2(v, t) = \left(1 - \frac{vm}{v_2}\right) < \Delta C_2(v, t) > + O(v^2).$$
 (58)

 $<\Delta C_2(v, t)>$ is approximately the average change in $C_2(v, t)$ between successive collisions given by

$$\langle \Delta C_2(v, t) \rangle \approx \int_{-\infty}^{t} e^{-\nu_m (t-t')} \nu_m dt' \left[C_2(v, t) - C_2(v, t') \right].$$
(59)

Proceeding in an analogous fashion we can get similar expressions for the $C_{\ell}(v, t)$ with $\ell > 2$. For $\ell > 2$ the analogues to equations (57) and (58) have a term going as γ^3 or higher in place of the γ^2 term. Thus, in all cases the requirement that the $C_{\ell}(v, t)$ with $\ell > 2$ be of order γ or higher, which is necessary for the validity of the generalized versions of equations (37), (1) and (44), is seen to reduce to the requirement that $(1 - \nu_m/\nu_\ell) < \Delta C_{\ell}(v, t) >$ be of order γ or higher for all $\ell \geq 2$.

In the fairly low energy range where one is justified in ignoring inelastic processes, the DeBroigle wave of the electron can be expected to be long compared to the range of molecular forces, in which case the scattering will be mostly s-wave scattering. Then $v \simeq v_0 \simeq v_m$ and the first term on the r.h.s. of equation (58) will be small. (This would not hold for molecules with appreciable static dipole moments.) However, separately we expect $\langle \Delta C_0(v, t) \rangle$ to be small, particularly for the steady state. When a steady state is obtained, we could separate $C_0(v, t)$ into a constant part, which would contribute nothing to $\langle \Delta C_0(v, t) \rangle$, and an oscillating part. Since the only agency tending to produce oscillatory motion is the electric field, we would expect that the oscillations

would be at one or more harmonics of the field frequency and that this part of C χ (v, t) and hence $<\Delta C \chi$ (v, t) > , would go as some power of γ .

FOOTNOTES AND REFERENCES

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